On Josephson effects in insulating spin systems

A. Schilling[a], H. Grundmann[a]

[a] *Physics Institute, University of Zürich, Winterthurerstrasse 190, CH-8057 Zürich*

Abstract

We discuss an experiment in which two magnetic insulators that both show a field-induced magnetic-ordering transition are weakly coupled to one another and are placed into an external magnetic field. If the respective magnetic states can be interpreted as phase coherent Bose-Einstein condensates of magnetic bosonic quasiparticles, one expects the occurrence of Josephson effects. For two identical systems, the resulting d.c. Josephson effect formally represents a constant quasiparticle Josephson current across the weak link, which turns out to be unobservable in an experiment. For magnetic insulators with different critical fields, a spontaneous alternating quasiparticle current develops with a leading oscillation frequency $\omega_{\text{a.c.}}$ that is determined by the difference between the critical fields. As a result of the coupling, additional sidebands appear in the energy spectrum of the coupled device that would be absent without phase coherence. We discuss the primary conditions for such an effect to take place and conclude that its detection can be feasible for a proper choice of compounds with suitable and realistic material parameters.

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1. Introduction

Quantum spin systems in solids have been a subject of intense research, both theoretically and experimentally. A number of such systems show magnetic-field induced phase transitions at zero temperature that have been interpreted as a Bose-Einstein condensation of magnetic bosonic quasiparticles [1–8].

In insulating dimerized spin-1/2 systems, for example, one expects a condensation of triplet bosonic quasiparticles ("triplons") above a certain critical field H_c [4, 5] where the energy difference between the ground-state singlet and the lowest excited triplet states vanishes due to the Zeeman splitting. In the language of magnetism, this condensation corresponds to a field-induced antiferromagnetic ordering that is associated with the appearance of a staggered magnetization [1, 3, 4]. However, the stability of such a condensate [9, 10] and even the applicability of the BEC concept [11–13] have been questioned.

Email address: aschilling@pysik.uzh.ch (A. Schilling)

Already known Bose-Einstein condensates (BEC) include superfluid helium [14, 15], dilute atomic gas clouds [16] and pumped exciton-polariton and magnon condensates [17–20]. Several experimental proofs for the existence of a macroscopic quantum state have been reported, e.g., from interference experiments [21–23], from the observation of vortices [24–26] and Josephson effects [27–32], and even superfluid properties can be ascribed to most of these systems [14, 15, 33–36]. Such hallmarks of a true BEC state are all based on the existence of a macroscopic phase-coherent state, but none of these has ever been observed in any insulating quantum spin system.

In this article we propose an experiment to probe the macroscopic phase coherence in insulating spin systems by observing the a.c. Josephson effect across a weak link. A successful detection of this effect would represent a direct proof for the existence of a macroscopic quantum state. We start with a discussion about the macroscopic wavefunction and the chemical potential of a triplon BEC in a magnetic insulator at zero temperature (sections 2 and 3). We then consider the Josephson equations for a system of two weakly coupled triplon BEC's (section 4) and show how the a.c. Josephson effect can be identified in principle. In section 5 we discuss several contraints for a system with realistic material parameters, and we finally conclude under realistic assumptions that such an experiment is indeed feasible.

2. The macroscopic wavefunction of a triplon BEC

The amplitude of the macroscopic wave function $\psi = \sqrt{n}e^{i\phi}$ describing a Bose-Einstein condensate of N triplons in a magnetic insulator at zero temperature is related to the density n, here defined as $n = N/N_d$ (with N_d the total number of dimers) so that $0 \leq n \leq 1$. To first approximation and near the critical field H_c, this density is proportional to the longitudinal magnetic moment $M_z = g\mu_B n N_d = g\mu_B N$ [5, 8], where g is the Landé g-factor and μ_B the Bohr magneton. As the equilibrium M_z is a well-defined quantity for a fixed value of the external magnetic field H in the thermodynamic limit, the triplon number $N = M_z/g\mu_B$ can be considered to be conserved.

The phase ϕ of the macroscopic wave function in magnetic insulators is closely related to (but not identical with) the angle φ of the transverse magnetic moments within the plane perpendicular to the main magnetic field H [7, 37, 38]. A plausibility argument for this fact is that the particle number N is canonically conjugate to the phase ϕ of the macroscopic wave function on the one hand, but also (via the proportionality of N to the longitudinal magnetic moment M_z and therefore to the associated component of the angular momentum L_z parallel to H) to the angle variable φ in the plane perpendicular to H on the other hand. In contrast to the time dependent global phase $\phi(t)$ of the macroscopic wavefunction, the angle φ between the transverse magnetic moments and the frame of reference given by the crystal lattice must be constant in time, because the Landau-Lifshitz-Gilbert equation does not allow for a perpetual spin precession of the staggered magnetization in the magnetic ground state. This apparent discrepancy can be easily resolved if one interprets the global phase of the macroscopic wave function as $\phi(t) = \varphi - \omega t$ (with ω provided by the time-dependent Schrödinger equation), i.e., the in-plane angle φ of the staggered magnetization corresponds to an undetermined phase constant ϕ_0. This interpretation of ϕ and φ ensures

that the expectation values of L_z and of the energy E per triplon are $\langle L_z \rangle_\psi = \hbar$ and $\langle E \rangle_\psi = \hbar\omega$, respectively.

In an axially symmetric spin system, φ must be able to take any arbitrary constant value between 0 and 2π [37], which is in certain contrast to experimental observations where φ seems to be locked to a material-specific value [39, 40]. We will address this issue of a violated axial symmetry later on in section 5.2. In the following we shall focus on the time evolution of the global phase $\phi(t)$ of the macroscopic wave function because its relation to the angle φ does not play any further important role here.

3. The chemical potential μ

A decisive prerequisite for an a.c. Josephson effect to take place between two BEC is the presence of a steady non-zero difference $\Delta\mu$ between the respective chemical potentials. In a condensate of magnetic quasiparticles in an insulating spin system, this chemical potential is usually taken as $\mu = g\mu_B\mu_0(H - H_c)$ [1, 2, 4, 5]. As in all BEC of weakly interacting light bosons in the dilute limit $n \ll 1$, a non-zero μ is related to n according to $n = \mu/v_0$. The interaction constant v_0 describes the repulsive hard-core interaction between the bosonic quasiparticles, and is in magnetic insulators determined by the finite inter-dimer interactions which result in the formation of dispersive energy bands of the triplet excitations [4]. It is important to emphasize that the appearance of a non-zero value of $\mu = dE/dN$ (with E the total energy of the condensate) is common to all interacting BEC that are treated within the Gross-Pitaevskii formalism [41, 42], and is not restricted to the magnetic systems under study here.

In the following we aim to bring two of such condensates into contact in order to investigate possible analogues to the d.c. and a.c. Josephson effects. Therefore it is indispensable to examine whether or not the above definition of the chemical potential μ is actually applicable to treat such an experiment correctly. If we consider, for example, two different magnetic insulators (α and β) with different critical magnetic fields $H_{c,\alpha}$ and $H_{c,\beta}$ that are placed into a common external magnetic field H, an apparently constant difference $\Delta\mu = \mu_\alpha - \mu_\beta = g\mu_0\mu_B(H_{c,\beta} - H_{c,\alpha})$ is maintained by the external magnetic field (see Fig. 1). This situation is formally equivalent to a device composed of two pieces of the same material with a single H_c, but placed into two different magnetic fields differing by ΔH. As we shall see later on, the necessary field gradient for the latter type of experiment to be successful is of the order of $\mu_0\Delta H \approx 0.5\,\mathrm{T}$ along a length of $\approx 0.5\,\mathrm{nm}$. This is technically out of reach, and we therefore do not consider this scenario any further.

This non-zero difference $\Delta\mu$ is not simply a result of choosing different reference points of the energy scale for the two condensates. Firstly, the difference in the critical fields $H_{c,j}$ ($j = \alpha$ or β) originally stems from a difference in microscopic inter- and intra-dimer coupling constants that determine the individual energy gaps, i.e. the energy separation of the respective ground-state singlet and the triplet states in zero magnetic field. To close these energy gaps, different external magnetic fields $H_{c,j}$ must be applied beyond which the respective ground-state triplons condense. On an absolute energy scale (with the zero point chosen for a state with isolated spin-1/2 particles in vacuum and in zero magnetic field) the

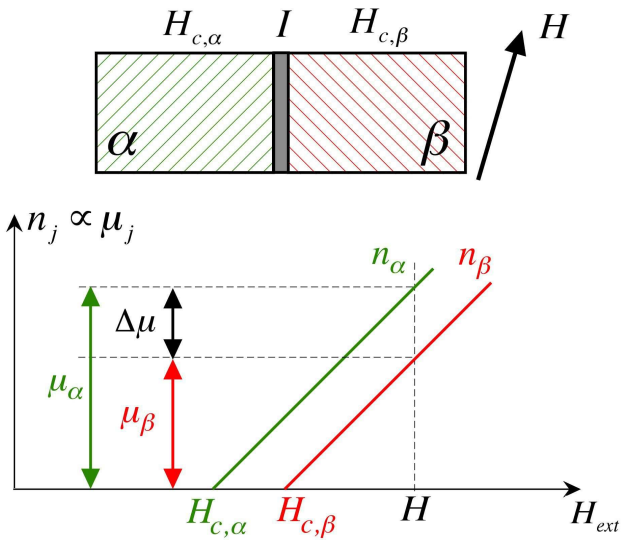

Figure 1: Sketch of an experiment in which two magnetic insulators with different critical fields $H_{c,j}$ ($j = \alpha, \beta$) are weakly coupled to one another through a magnetically inert spacer layer I and are placed into an external magnetic field $H > H_{c,j}$. The respective condensate densities n_j and the chemical potentials μ_j vary approximately linearly with H.

condensation occurs at the energy level of the respective singlet states, i.e., at $E_j = -3/4J_j$ per dimer, where J_j denote the respective intra-dimer coupling energies. Secondly and most importantly, the relevant quantity that enters the problem of treating the Josephson effect is the relative energy gain (loss) dE/dN upon creation (or annihilation) of one triplon quasiparticle to (or from) the condensate, from (or into) the singlet sea, which is actually $n_j v_{0,j} = \mu_j = g_j \mu_B \mu_0 (H - H_{c,j})$ per dimer[1] [3–5, 7]. Therefore, the μ_j (and along with them the condensate densities $n_j = \mu_j / v_{0,j}$) are indeed different in a common external magnetic field that exceeds both $H_{c,\alpha}$ and $H_{c,\beta}$ (see Fig. 1), and $\Delta\mu$ can then be considered as the analogue to an external voltage controlling the difference between the chemical potentials in a superconducting Josephson junction.

4. Josephson effects

4.1. The Josephson equations

We now apply this concept to a system of two dimerized spin systems at zero temperature and with different critical fields $H_{c,\beta} > H_{c,\alpha}$ beyond which the respective magnetic quasiparticles are supposed to condense. We initially assume a perfect axial symmetry of the two magnetic systems with respect to the direction of the external magnetic field, but we will discuss the case of violated axial symmetry later on in section 5.2. The boundary

[1] This result for μ can be illustrated by identifying E with the energy of the magnetized system in a field H, $E = \mu_0 \int_{H_c}^{H} M_z(H')\,\mathrm{d}H'$ with $M_z = g\mu_B N$ and $N = g\mu_B\mu_0\,(H - H_c)\,N_d/v_0$, so that $E(N) = N^2 v_0 / 2N_d$ and $\mu = \mathrm{d}E/\mathrm{d}N = nv_0$.

layers of the two systems are assumed to be weakly coupled to one another, and we place the device into a magnetic field $H > H_{c,j}$ (see Fig. 1). For simplicity, we also assume $v_{0,\alpha} \approx v_{0,\beta} = v_0$ and $g_\alpha \approx g_\beta = g$, which is not essential for the main conclusion of our consideration, however.

In an approach introduced by Feynman [43] to explain the Josephson effects across a weak link between two superconductors one considers the macroscopic wave functions $\psi_j = \sqrt{n_j} e^{i\phi_j}$ on opposite sides of the junction, and treats the weak coupling between them according to $i\hbar \frac{\partial}{\partial t} \psi_{\alpha;\beta} = \mu_{\alpha;\beta} \psi_{\alpha;\beta} + K \psi_{\beta;\alpha}$, where $K \ll \mu_j$ is a phenomenological coupling constant. The resulting differential equations for the number of particles n_j occupying the respective macroscopic quantum states and for the corresponding phase difference $\Delta\phi = \phi_\beta - \phi_\alpha$ become

$$\frac{\partial n_\alpha}{\partial t} = -\frac{\partial n_\beta}{\partial t} = \frac{2K}{\hbar} \sqrt{n_\alpha n_\beta} \sin(\Delta\phi) , \tag{1a}$$

$$\frac{\partial \Delta\phi}{\partial t} = \frac{\mu_\alpha - \mu_\beta}{\hbar} - \frac{K}{\hbar} \frac{n_\alpha - n_\beta}{\sqrt{n_\alpha n_\beta}} \cos(\Delta\phi) . \tag{1b}$$

For $H_{c,\beta} > H_{c,\alpha}$, the solution of Eq. 1b in the weak-coupling limit $K \ll \mu_\beta$ is, to first approximation, $\Delta\phi \approx \Delta\phi_0 + \omega_{\text{a.c.}} t$ with $\omega_{\text{a.c.}} \approx \Delta\mu/\hbar$ and an undetermined constant phase difference $\Delta\phi_0$. The resulting variations in $n_j(t)$ are very small, i.e, by a factor $K/\Delta\mu$ smaller than their time-averaged values.

4.2. The d.c. and a.c. Josephson effects

In the case of identical systems ($\Delta\mu = 0$ and $n_\alpha = n_\beta$, see Fig. 2a) a constant $\Delta\phi(t) = \Delta\phi_0 = \phi_{0,\beta} - \phi_{0,\alpha}$ would represent a constant quasiparticle current $\partial n_\alpha/\partial t$ across the weak link (d.c. Josephson effect). As the constants $\phi_{0,j}$ correspond to the in-plane angles φ_j of the respective transverse magnetic moments that will tend to align in the coupled device, we expect $\Delta\phi_0 = 0$ and therefore $\partial n_j/\partial t = 0$.

In a device with two magnetic insulators with different critical fields, however, $\Delta\mu \neq 0$ and therefore $\omega_{\text{a.c.}} \neq 0$ (Fig. 2c). As one would expect it from the analogy with superconductors and superfluids, the resulting variation $\partial n_\alpha/\partial t$ given by Eq. 1a represents an oscillation in time with the field-independent leading frequency $\omega_{\text{a.c.}}$ (a.c. Josephson effect).

The analogous situation in a superconducting Josephson device leads to the appearance of Josephson electrical currents. If connected to an external charge reservoir and fixing $\Delta\mu$ to a constant value, the net charge-carrier density remains constant despite a nonzero $\partial n_\alpha/\partial t$, but a measurable electrical current proportional to $\partial n_\alpha/\partial t$ flows from the charge reservoir and the attached leads through the whole device [43].

With a junction composed of insulating spin systems it is the external magnetic field that maintains the difference between the chemical potentials to a certain value. This field fully penetrates the whole sample volume, and it therefore entirely "short-circuits" each of the two spin systems separately, thereby keeping the respective total numbers of quasiparticles n_j constant. As a consequence, there is no directional macroscopic quasiparticle current flowing *within* the two individual branches on both sides of the device. One can think

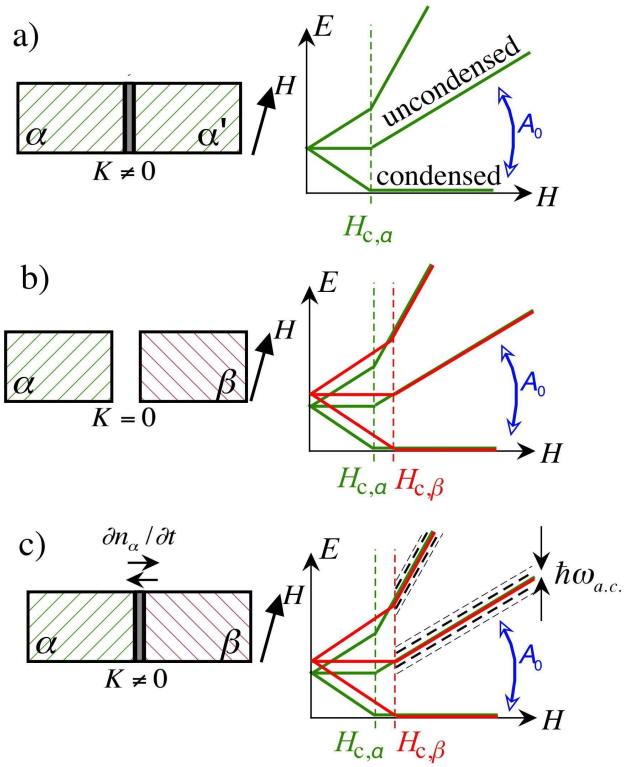

Figure 2: Sketch of the expected energy scheme (right panels) for a) two identical weakly coupled magnetic insulators, b) two uncoupled magnetic insulators with different critical fields, and c) two weakly coupled magnetic insulators with different critical fields (a.c. Josephson effect), all for $H > H_{c,j}$ and measured relative to the respective ground-state energies. In the case c), the ESR-active mode A_0 should split by a detectable amount $\hbar\omega_{\mathrm{a.c}}$ (see text). Arrows in the left panels symbolize the quasiparticle currents related to $\partial n_\alpha/\partial t$ that can be ascribed to the spacer region (see text).

of the magnetic field as a quasiparticle source (or sink) that replaces (or removes) those quasiparticles that are crossing the weak link. It is only by virtue of the phase difference $\Delta\phi$ between the two materials that a quasiparticle current can still be formally ascribed to the magnetically inert spacer region (see Fig. 2c), but it may be impossible to detect it directly in an experiment.

4.3. Experimental manifestation of the a.c. Josephson effect

To second-order approximation ($K \neq 0$ with $H_{c,\alpha} \neq H_{c,\beta}$ and therefore $n_\alpha \neq n_\beta$), Eq. 1b denotes a narrowband frequency modulation of the phases

$$\phi_{\alpha;\beta}(t) \approx \phi_{0,\alpha;\beta} - \frac{\mu_{\alpha;\beta}}{\hbar}t - \frac{K}{\Delta\mu}\sqrt{\frac{n_{\beta;\alpha}}{n_{\alpha;\beta}}}\sin(\omega_{\mathrm{a.c.}}t + \Delta\phi_0), \tag{2}$$

with a modulation frequency $\omega_{\mathrm{a.c.}}$ and a modulation index of the order of $K/\Delta\mu$. This modulation of $\phi_j(t)$ has profound consequences on the macroscopic wave functions $\psi_j = \sqrt{n_j}e^{i\phi_j}$ describing the two condensates, because equally spaced sidebands should appear in the energy spectrum of the coupled device. This dynamic effect should manifest itself

in a corresponding splitting of all energies that are associated with transitions from the (condensed) ground state to (uncondensed) excited states (see Fig. 2c). Such a splitting is absent for two uncoupled magnetic insulators with different critical fields (Fig. 2b) and, of course, also in a scenario where no macroscopic phase coherence is present at all. Therefore a successful experiment in a coupled system that can probe the occurrence of sidebands with separation $\hbar\omega_{\mathrm{a.c.}}$ would represent a very strong experimental support for the existence of a state with macroscopic phase coherence.

To be more specific, a high-resolution electron-spin resonance (ESR) measurement of the transitions between the ground state considered here, and the nearest excited triplet states (referred to as A_0 in Ref. [44] and $E_0(Q)$ in Ref. [45]), or a one-magnon Raman experiment on the so-called $E_-(Q)$-mode [46], should reveal a characteristic splitting of the corresponding modes by $\hbar\omega_{\mathrm{a.c.}}$ which is related to the a.c. Josephson effect (see Fig. 2c). This is a central result of this work, and we shall discuss in the following whether or not the proposed a.c. Josephson effect can be observed in a device with realistic material parameters.

5. Constraints in real systems

5.1. The lifetime of phase coherence

We may expect a successful experiment only if the separation $\hbar\omega_{\mathrm{a.c.}}$ between associated sidebands in the energy spectrum is comparable to or larger than their linewidth which can be related to the inverse lifetime τ_{pc}^{-1} of phase coherence and to the inverse lifetime τ_{qp}^{-1} of the magnetic quasiparticles involved, i.e., $\omega_{\mathrm{a.c.}} \gtrsim max(\tau_{pc}^{-1}, \tau_{qp}^{-1})$. The lifetime τ_{pc} has, to the best of our knowledge, not yet been estimated for triplon BEC's, but we may initially compare it to τ_{qp}, i.e., that of the $S = 1$ quasiparticles. Scattering processes on magnons, phonons or impurities can be important limiting factors, but there is no fundamental principle that would restrict it to inaccessibly short time scales at low enough temperatures. Corresponding values for the quasiparticle lifetime τ_{qp} ranging from $> 10^{-11}\,\mathrm{s}$ up to $\approx 5 \times 10^{-11}\,\mathrm{s}$ can be inferred from precise inelastic neutron-scattering measurements of the magnon linewidth in the spin-dimerized compounds $TlCuCl_3$ [47] and $(C_4H_12N_2)Cu_2Cl_6$ [48], respectively, taken at low temperatures for quasiparticles that are not part of the condensate, and from the ESR linewidth of the A_0 mode observed in $TlCuCl_3$ [44].

It is conceivable that the actual lifetime τ_{pc} of phase coherence in the BEC is even considerably longer than that of its constituting particles τ_{qp}, e.g., thanks to the formation of energetic barriers of topologic origin that may lead to a strong suppression of dissipation, i.e., to the occurrence of true spin superfluidity [37]. It is worth mentioning here that the a.c. Josephson oscillations that have been seen in an experiment on an exciton-polariton Josephson junction were clearly observable although the oscillation frequency ($\omega_{\mathrm{a.c.}} \approx 4 \times 10^{11}\,\mathrm{s}^{-1}$) was very near the inverse lifetime of the respective bosonic quasiparticles ($\tau_{qp}^{-1} \approx 3 \times 10^{11}\,\mathrm{s}^{-1}$) [32], but still much larger than the inverse of the surprisingly long measured phase-coherence time ($\tau_{pc}^{-1} \approx 7 \times 10^{9}\,\mathrm{s}^{-1}$ [49], i.e., $\tau_{pc} >> \tau_{qp}$). To be on the safe side we conservatively choose $\omega_{\mathrm{a.c.}} \approx 10^{11}\,\mathrm{s}^{-1} > \tau_{qp}^{-1}$ for our proposed experiment, which corresponds to $\mu_0(H_{c,\beta} - H_{c,\alpha}) \approx 0.5\,\mathrm{T}$.

5.2. Violated axial symmetry

So far we have considered only perfectly axially symmetric materials. Any violation of axial symmetry of the magnetic exchange interaction leads to the occurrence of an anisotropy gap Δ in the energy spectrum $E(k)$ [54], thereby lifting the Goldstone linearity for $k \to 0$, fixing the phases $\phi_{0,j}$ to constant values [3, 9] and leading to well-defined magnetic structures with in-plane angles φ_j that are locked to the crystal lattice [39, 40]. This gap has been estimated to $\Delta(H) = \sqrt{8\tilde{\gamma}\mu(H)}$, where $\tilde{\gamma} > 0$ is a measure for the exchange-interaction anisotropy in the plane perpendicular to H [54]. For experimental energies larger than Δ, however, a quasi-linear Goldstone mode is recovered [6], the effects of axial anisotropy are expected to be smeared out on short enough time scales, and we can ascribe an upper limit $\tau_\Delta = \hbar/\Delta$ to the lifetime of phase coherence [9] so that we must additionally require $\omega_{\mathrm{a.c.}} > \tau_\Delta^{-1}$.

5.3. The weak link

In a real experiment, $\Delta\mu = g\mu_0\mu_B(H_{c,\beta} - H_{c,\alpha})$ and the coupling constant K are determined by the junction properties. The dimensions of the weak link should not significantly exceed the healing length of the condensate (the analogue to the coherence length in superconductors). For weakly interacting Bose gases, this healing length is $\xi = \hbar/\sqrt{2nv_0m^*}$ (where $m*$ is the effective mass of the quasiparticles) [50] and it diverges as the temperature is increased towards the Bose-Einstein condensation temperature where n vanishes.

The simplest version of such a weak link may consist of a slit that is narrow enough to allow for a small but finite magnetic coupling and therefore for a tunnelling of magnetic quasiparticles. Such a junction might also be manufactured by the layered growth of a magnetic insulator (α, with $H_{c,\alpha}$) containing a certain species of magnetic ions forming spin-1/2 dimers, followed by one or a few layers of an isostructural compound with nonmagnetic ions, and by further adjacent layers of another isostructural magnetic insulator (β, with $H_{c,\beta} \neq H_{c,\alpha}$).

5.4. A realistic toy model: (Ba,Sr)$_3$Cr$_2$O$_8$

To obtain numeric values, we consider a model device composed of $Ba_3Cr_2O_8$ (α) with $\mu_0 H_{c,\alpha} \approx 12\,\mathrm{T}$ [40], separated by nonmagnetic isostructural $Ba_3V_2O_8$ [51] from a compound $Ba_{3-x}Sr_xCr_2O_8$ (β) [52][2] with x chosen in such a way to achieve a larger $\mu_0 H_{c,\beta}$, but still much smaller than $\mu_0 H_{c2} = 30\,\mathrm{T}$ of fully stoichiometric $Sr_3Cr_2O_8$ with $x = 1$ [52]. If we achieve a $\mu_0 H_{c,\beta} \approx 12.5\,\mathrm{T}$, the expected characteristic frequency becomes $\omega_{\mathrm{a.c.}} \approx 10^{11}\,\mathrm{s}^{-1}$. With an external magnetic field $\mu_0 H \approx 13\,\mathrm{T}$, and taking $v_0/k_B \approx 8.7\,\mathrm{K}$ with $g \approx 2$ [53] we have $n_\alpha \approx 0.15$ and $n_\beta \approx 0.08$. The choice of a small $H - H_{c,j}$ (i.e., $\mu_j \ll v_0$) is primarily necessary to fulfill the dilute-limit condition $n_j \ll 1$, but it may not be crucial for observing the essence of the predicted effect. With $m^* \approx 1.5 \times 10^{-27}\,\mathrm{kg}$ and a mean distance $d \approx 0.6\,\mathrm{nm}$ between the dimers [40, 53], the resulting values for the healing lengths at $T = 0$ are small

[2] We have already verified the existence of the solid-solution series $Ba_{3-x}Sr_xCr_2O_8$ for polycrystalline samples, but we have no further information about their magnetic properties.

($\xi_\alpha \approx 0.4\,\mathrm{nm}$ and $\xi_\beta \approx 0.6\,\mathrm{nm} \approx d$), but a very narrow slit or a weak link composed of one or only a few unit cells of the nonmagnetic compound could be adequate. If still larger values of ξ_j are required, n_j can be diminished by further reducing $\Delta\mu$ and μ_β. Alternatively, the experiment can be performed near the condensation temperature as it has been done in superfluid ^{4}He to make the a.c. Josephson effect observable [27], possibly at the cost of reducing the lifetime τ_{qp} of the triplon quasiparticles.

With the material parameters used for the experiment described above and $\omega_{\mathrm{a.c.}} \approx 10^{11}\,\mathrm{s}^{-1}$, $\tilde{\gamma}$ should be below $\approx 4\,\mu\mathrm{eV}$. The exchange-interaction anisotropy has been estimated to $\tilde{\gamma} \approx 16 - 30\,\mu\mathrm{eV}$ in $TlCuCl_3$ [9, 55], but to only $\tilde{\gamma} \approx 1\,\mu\mathrm{eV} < 4\,\mu\mathrm{eV}$ in $BaCuSi_2O_6$ [56] and particularly in $Ba_3Cr_2O_8$ [40], so that the proposed device would fulfil all of the requirements listed above.

6. Concluding remarks

To conclude these numeric estimates we want to state that other material properties, such as the presence of a strong Dzyaloshinski-Moriya interaction that we have not considered here, may still inhibit the formation of a phase coherent condensate [54] and therefore make the observation of the a.c. Josephson effect impossible. However, we classify such factors as not intrinsic to the problem, and a proper choice of compounds with suitable material parameters should make the generation and the observation of the a.c. Josephson effect in magnetic insulators feasible in principle. Although we have chosen dimerized spin systems operating near their lower critical fields as a model for the present consideration, all the above arguments should also hold near the respective saturation fields as a consequence of the particle-hole symmetry of the problem [4], and for all other types of insulating spin systems that are supposed to show a field-induced BEC of magnetic quasiparticles. It is conceivable that Josephson-like phenomena can even naturally occur in certain quantum magnets, e.g., in a system showing an intrinsic modulation of the boson density (and therefore of the chemical potential) due to inequivalent planes hosting the triplon condensate. Such a situation has been reported for $BaCuSi_2O_8$, in which the average boson density strongly varies along the c-axis [57, 58].

We finally mention here an interesting analogy between the case of an ESR measurement on weakly coupled magnetic insulators as discussed above, and corresponding microwave stimulated experiments on ferromagnetic films which are separated by normal-metal spacers [59]. In both cases, there exists a dynamic coupling between two different materials through a magnetically inert spacer layer, thereby altering the dynamics in both systems as compared to the uncoupled situation.

In summary, we suggest that two weakly linked magnetic insulators with different critical fields $H_{c,j}$ that are placed into a suitably chosen external magnetic field should show an altered energy spectrum as compared to the uncoupled limit, with additional sidebands that are separated by $\hbar\omega_{\mathrm{a.c.}} = g\mu_B\mu_0(H_{c,\beta} - H_{c,\alpha})$. These sidebands are a manifestation of the a.c. Josephson effect and can be tested, for example, in a high-resolution ESR experiment. Assuming realistic material parameters and considering several constraints, we conclude that this effect can indeed take place in a real system. A corresponding successful

experiment would represent a very strong experimental support for the existence of a state with macroscopic phase coherence.

7. Acknowlegdements

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References

[1] T. Matsubara and H. Matsuda, Prog. Theor. Phys. **16** (1956) 569.
[2] E. G. Batyev and L. S. Braginskii, Sov. Phys. JETP **60** (1984) 781.
[3] I. Affleck, Phys. Rev. B **43** (1991) 3215.
[4] T. Giamarchi and A. M. Tsvelik, Phys. Rev. B **59** (1999) 11398.
[5] T. Nikuni, M. Oshikawa, A. Oosawa and H. Tanaka, Phys. Rev. Lett. **84** (2000) 5868.
[6] C. Rüegg, N. Cavadini, A. Furrer, H.-U. Güdel, K. Krämer, H. Mutka, A. Wildes, K. Habicht and P. Vorderwisch, Nature **423** (2003) 62.
[7] T. Giamarchi, C. Rüegg and O. Tcherniyshyov, Nature Phys. **4** (2008) 198.
[8] M. Matsumoto, B. Normand, T. M. Rice and M. Sigrist, Phys. Rev. B **69** (2004) 054423.
[9] R. Dell'Amore, A. Schilling and K. Krämer, Phys. Rev. B **79** (2009) 014438.
[10] A. Rakhimov, E. Y. Sherman and C. K. Kim, Phys. Rev. B **81** (2010) 020407.
[11] V. M. Kalita and V. M. Loktev, JETP Lett. **91** (2010) 183.
[12] Y. M. Bunkov and G. E. Volovik, arXiv:1003.4889v1.
[13] D. L. Mills, Phys. Rev. Lett. **98** (2007) 039701.
[14] J. F. Allen and A. D. Misener, Nature **141** (1938) 75.
[15] P. Kapitza, Nature **141** (1938) 74.
[16] M. H. Anderson, J. R. Ensher, M. R. Matthews, C. E. Wieman and E. A. Cornell, Science **269** (1995) 198.
[17] S. O. Demokritov, V. E. Demidov, O. Dzyapko, G. A. Melkov, A. A. Serga, B. Hillebrands and A. N. Slavin, Nature **443** (2006) 430.
[18] V. E. Demidov, O. Dzyapko, S. O. Demokritov, G. A. Melkov and A. N. Slavin, Phys. Rev. Lett. **100** (2008) 047205.
[19] H. Deng, G. Weihs, C. Santori, J. Bloch and Y. Yamamoto, Science **298** (2002) 199.
[20] J. Kasprazak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J. M. J. Keeling, F. M. Marchetti, M. H. Szymanska, R. André, J. L. Staehli, V. Savona, P. B. Littlewood, B. Deveaud and L. S. Dang, Nature **443** (2006) 409.
[21] E. Hoskinson, Y. Sato and R. Packard, Phys. Rev. B **74** (2006) 100509.
[22] R.W. Simmonds, A. Marchenkov, E. Hoskinson, J. C. Davis and R. E. Packard, Nature **412** (2001) 55.
[23] M. R. Andrews, C. G. Townsend, H.-J. Miesner, D. S. Durfee, D. M. Kurn and W. Ketterle, Science **275** (1997) 637.
[24] H. E. Hall and W. F. Vinen, Proceedings of the Royal Society A **238** (1956) 204.
[25] M. R. Matthews, B. P. Anderson, P. C. Haljan, D. S. Hall, C. E. Wieman and E. A. Cornell, Phys. Rev. Lett. **83** (1999) 2498.
[26] K. G. Lagoudakis, M. Wouters, M. Richard, A. Baas, I. Carusotto, R. André, L. S. Dang and B. Deveaud-Plédran, Nature Phys. **4** (2008) 706.
[27] K. Sukhatme, Y. Mukharsky, T. Chui and D. Pearson, Nature **411** (2001) 280.
[28] A. S. Borovik-Romanov, Y. M. Bun'kov, A. deVaard, V. V. Dmitriev, V. Matrotsieva, Y. M. Mukharskii and D. A. Sergatskov, JETP Lett. **47** (1988) 478.
[29] S. V. Pereverzev, A. Loshak, S. Backhaus, J. C. Davis and R. E. Packard, Nature **388** (1997) 449.

[30] B. P. Anderson and M. A. Kasevich, Science **282** (1998) 1686.
[31] S. Levy, E. Lahoud, I. Shomroni and J. Steinhauer, Nature **449** (2007) 579.
[32] K. G. Lagoudakis, B. Pietka, M. Wouters, R. André and B. Deveaud-Plédran, Phys. Rev. Lett. **105** (2010) 120403.
[33] V. P. Peshkov, J. Physics (Moscow) **8** (1944) 381.
[34] A. S. Borovik-Romanov, Y. M. Bun'kov, V. V. Dmitriev and Y. M. Mukharskii, JETP Lett. **40** (1984) 1033.
[35] C. Raman, M. Köhl, R. Onofrio, D. S. Durfee, C. E. Kuklewicz, Z. Hadzibabic and W. Ketterle, Phys. Rev. Lett. **83** (1999) 2502.
[36] A. Amo, D. Sanvitto, F. P. Laussy, D. Ballarini, E. del Valle, M. D. Martin, A. Lemaître, J. Bloch, D. N. Krizhanovskii, M. S. Skolnick, C. Tejedor and Viña L., Nature **457** (2009) 291.
[37] E. B. Sonin, Adv. Phys. **59** (2010) 181.
[38] F. S. Nogueira and K.-H. Bennemann, Europhys. Lett. **67** (2004) 620.
[39] H. Tanaka, A. Oosawa, T. Kato, H. Uekusa, Y. Ohashi, K. Kakurai and A. Hoser A., J. Phys. Soc. Jpn. **70** (2001) 939.
[40] M. Kofu, H. Ueda, H. Nojiri, Y. Oshima, T. Zenmoto, K. C. Rule, S. Gerischer, B. Lake, C. D. Batista, Y. Ueda and S.-H. Lee, Phys. Rev. Lett. **102** (2009) 177204.
[41] L. P. Pitaevskii and S. Stringari, Bose-Einstein Condensation, Oxford Science Publications, 2003.
[42] R. Ozeri, N. Katz, J. Steinhauer and N. Davidson, Rev. Mod. Phys. **77** (2005) 187.
[43] R. P. Feynman, R. B. Leighton and M. Sands, The Feynman Lectures on Physics Vol. III, Addison-Wesley, 1965.
[44] S. Kimura, M. Hagiwara, H. Tanaka, A. Kolezhuk and K. Kindo, J. Magn. Magn. Mater. **310** (2007) 1218.
[45] M. Matsumoto, T. Shoji and M. Koga M., J. Phys. Soc. Jpn. **77** (2008) 074712.
[46] H. Kuroe, K. Kusakabe, A. Oosawa, T. Sekine, F. Yamada, H. Tanaka and M. Matsumoto, Phys. Rev. B **77** (2008) 134420.
[47] C. Rüegg, B. Normand, M. Matsumoto, C. Niedermayer, A. Furrer, K. W. Krämer, H.-U. Güdel, P. Bourges, Y. Sidis and H. Mutka, Phys. Rev. Lett. **95** (2005) 267201.
[48] M. B. Stone, C. Broholm, D. H. Reich, P. Schiffer, O. Tchernyshyov, P. Vorderwisch and N. Harrison, New J. Phys. **9** (2007) 31.
[49] A. P. D. Love, D. N. Krizhanovskii, D. M. Whittaker, R. Bouchekioua, D. Sanvitto, S. A. Rizeiqi, R. Bradley, M. S. Skolnick, P. R. Eastham, R. André and L. S. Dang, Phys. Rev. Lett. **101** (2008) 067404.
[50] N. N. Bogoliubov, J. Phys. (USSR) **11** (1947) 23.
[51] P. Süsse and M. J. Buerger, Z. Kristallogr. **131** (1970) 161.
[52] A. A. Aczel, Y. Kohama, C. Marcenat, F. Weickert, M. Jaime, O. E. Ayala-Valenzuela, R. D. McDonald, S. D. Selesnic, H. A. Dabkowska and G. M. Luke, Phys. Rev. Lett. **103** (2009) 207203.
[53] T. Dodds, B.-J. Yang and Y. B. Kim, Phys. Rev. B **81** (2010) 054412.
[54] J. Sirker, A. Weisse and O. P. Sushkov, J. Phys. Soc. Jpn. **74** (2005) Suppl. 129.
[55] A. K. Kolezhuk, V. N. Glazkov, H. Tanaka and A. Oosawa A., Phys. Rev. B **70** (2004) 020403.
[56] S. E. Sebastian, P. Tanedo, P. A. Goddard, S.-C. Lee, A. Wilson, S. Kim, S. Cox, R. D. McDonald, S. Hill, N. Harrison, C. D. Batista and I. R. Fisher, Phys. Rev. B **74** (2006) 180401.
[57] S. Krämer, R. Stern, M. Horvatić, C. Berthier, T. Kimura, and I. R. Fisher , Phys. Rev. B **76** (2007) 100406(R).
[58] N. Laflorencie and F. Mila, Phys. Rev. Lett. **102** (2009) 060602.
[59] B. Heinrich, Y. Tserkovnyak, G. Woltersdorf, A. Brataas, R. Urban and G. E. W. Bauer, Phys. Rev. Lett. **90** (2003) 187601.